

# Biodegradable Paints for Daytime Radiative Cooling

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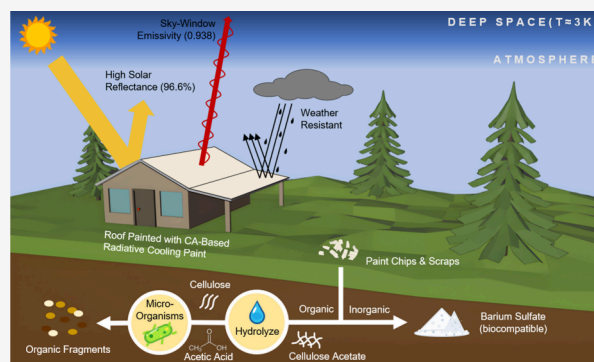
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Supporting Information

**ABSTRACT:** Radiative cooling technologies help alleviate global warming and the urban heat island effect by reducing the energy load of conventional air conditioning. However, most radiative cooling materials contain nonbiodegradable components that decompose into microplastics and threaten the environment. To address this issue, this work develops a biodegradable radiative cooling paint, experimentally demonstrated to maintain an average surface temperature of 2–3 °C below ambient temperature and around 5 °C below commercial white paint under high solar irradiation ( $>800 \text{ W m}^{-2}$ ). Fabricated samples showed a high solar reflectance of 96.6% and sky window emissivity of 0.938. The binder demonstrated complete biodegradation within 3 months in a compost environment. TRNSYS simulation shows high energy savings across the US, with over 400 kWh of electricity saved in hot and dry climates such as Phoenix, AZ. With high UV resistance, mechanical durability, and self-cleaning capabilities, the biodegradable radiative cooling paint is suitable for large-scale deployment.

**KEYWORDS:** radiative cooling, biodegradable paint, cellulose acetate, barium sulfate, nanoporous composites, energy savings



In the United States, air conditioners account for approximately 6% of all electricity generated, underscoring the urgency for alternative cooling methods.<sup>1</sup> Radiative cooling stands as a pivotal innovation within the realm of sustainable technologies, given its potential to reduce energy consumption in cooling systems significantly.<sup>2</sup> Radiative cooling solutions exploit the atmospheric transparency within the “sky window” (8 to 13  $\mu\text{m}$  wavelength range), which facilitates the direct dissipation of thermal radiation from surfaces to outer space without the need for energy input. However, the integration of radiative cooling materials into practical applications faces several challenges, including the complexity of synthesizing materials with intricate structures, extensive chemical usage, the inclusion of metallic layers, and poor adaptability across different surface morphologies.<sup>3–9</sup> Therefore, there is a critical need to develop high-performance and scalable radiative cooling solutions. Among many options, paint is one of the most compatible and robust form factors.

Li et al. demonstrated that barium sulfate ( $\text{BaSO}_4$ ) serves as an effective pigment.<sup>10</sup> When used in high concentrations within a paint matrix in an acrylic base,  $\text{BaSO}_4$ , as well as calcium carbonate ( $\text{CaCO}_3$ ), produces a coating with thicknesses ranging from 200 to 400  $\mu\text{m}$ . This paint not only exhibits outstanding properties such as ultrahigh solar reflectance and high emissivity within the sky window but also achieves full daytime cooling below ambient temperatures.<sup>10,11</sup> Furthermore, the use of core-shell particle technology has shown promise in enhancing radiative cooling performance using fillers such as silica, zinc oxide, and

acrylic.<sup>12</sup> Other research has explored the use of wide bandgap nanoparticle paints that also prioritize high filler concentrations to optimize radiative cooling efficacy. Despite achieving impressive cooling capabilities, these coatings have a thickness of up to 1 mm, which is thicker than typical paints and presents application challenges that must be overcome.<sup>13</sup> One recent advance in this area is the development of hexagonal boron nitride (hBN) paint, which achieves robust daytime sub-ambient cooling with a minimal coating thickness of 150  $\mu\text{m}$  using large bandgap fillers and a diversified particle size distribution.<sup>14</sup>

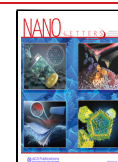
However, most carrier matrices of the radiative cooling paints contain one or more nonbiocompatible or nonbiodegradable components. They will be broken down to microplastics and could enter food chains at the end of their life to threaten the environment and associated dependents.<sup>15–21</sup> Particularly, the binders, responsible for carrying photonic particles in radiative cooling paint, are not decomposed by nature and stay in the ecosystem for hundreds of years. EA Switzerland projects 1.9 million tons of microplastic emissions into the ocean as a direct result of paint/coating waste, accounting for 40% of all plastic waste

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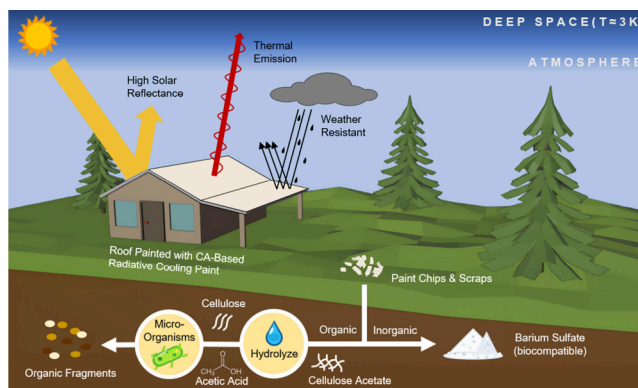
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influx into the ecosystem.<sup>22</sup> Therefore, several challenging but highly desirable advances are needed to push radiative cooling to eco-friendly and scalable applications: (1) identify components in these paints that support a full eco-friendly and nontoxic decomposition, (2) develop synthesis processes that avoid irritating chemicals and harmful waste, and (3) simultaneously achieve durability and biodegradability even when radiative coolers are exposed to sunlight, dust, rain, and pollution, which deteriorate the exposed surface over time. For that purpose, the fibrillated cellulose matrix harvested from renewable resources is technologically appealing for its sustainability potential and its innate radiative cooling capabilities.

Although biodegradable radiative cooling materials exist in alternative forms, e.g., cooling wood,<sup>23</sup> cooling textiles,<sup>24–30</sup> and cooling paper,<sup>31</sup> their cooling performance is usually limited due to unavoidable UV absorption (10–40%) of their polymer composition; hence it is desirable to create paint forms with enhanced performance. Nevertheless, the cellulose components in the above examples are demonstrated as a potential ingredient of radiative cooling paint for photonic scattering in most solar spectrum and innately high infrared (IR) absorption. Therefore, incorporating the respective benefits of dielectric nanocomposites and a cellulose matrix is ideal to achieve desirable optical properties for a wide range of radiative cooling applications. Recent studies have incorporated BaSO<sub>4</sub> into cellulose-derived radiative cooling systems, including BaSO<sub>4</sub>–acrylic coatings on cellulose substrates<sup>32</sup> and natural pulp–BaSO<sub>4</sub> composite coatings.<sup>33</sup> However, these works respectively use cellulose as a substrate/support layer or as a fibrous pulp matrix, rather than as a continuous biodegradable binder replacing acrylic in paint formulations. Native cellulose and pulp are difficult to process as conventional paint binders because of their limited solubility and dispersibility in common solvents, which originates from strong inter- and intramolecular hydrogen bonding. Therefore, a biodegradable binder that is more closely suited to the conventional processing and application requirements of paints is desirable.

In this work, we have designed and fabricated BaSO<sub>4</sub>–cellulose acetate (BaSO<sub>4</sub>–CA) paints. The inherently highly porous structure of cellulose acetate, used as a paint binder, enables the diffuse scattering of visible and near-infrared (NIR) radiation. The resultant composite has experimentally demonstrated high solar reflectance (96.6%) and sky window emissivity (0.938). As illustrated in Figure 1, when painted on a roof, the biodegradable radiative cooling paint reflects incoming sunlight while simultaneously emits heat between the wavelength of 8 and 13  $\mu\text{m}$ . Outdoor experiments of this composition have shown a maximum cooling effect of 9.5 °C below ambient temperature on the sample's surface. Moreover, the developed formulation exhibits wear resistance, UV resistance, and water resistance, aligning well with industry benchmarks. Critically, cellulose acetate enables the biodegradability of the polymeric binder within the paint, eliminating persistent polymeric residues in the environment. At the end of its life cycle, it completely biodegrades, breaking down into organic fragments when it enters the soil environment as chips or powders. In this environment, enzymes secreted by microorganisms such as bacteria and fungi cleave acetyl groups, reversing the synthesis of cellulose acetate. This breakdown results in the formation of cellulose and acetic acid. The microorganisms then consume the broken-down cellulose



**Figure 1.** Principle of cellulose acetate (CA) based radiative cooling paint. Schematic of functional principles and biodegradation process of CA–BaSO<sub>4</sub> radiative cooling paint, including mechanical and chemical breakdowns.

acetate fragments as a carbon source, converting them into simpler substances like water, carbon dioxide, and biomass. The presence of organic carbon matter serves as a rich source of energy and nutrients for microorganisms, which metabolize cellulose acetate to generate enzymes that further accelerate its decomposition. To demonstrate this, this work directly compares the compost degradation behavior of BaSO<sub>4</sub>–CA paint with that of an analogous BaSO<sub>4</sub>–acrylic radiative cooling paint, demonstrating degradation of the CA-containing binder phase while the acrylic-bound control remains persistent. The results of this work establish and validate a biodegradable radiative cooling paint with high cooling and durability performance during its lifetime while reducing binder-derived plastic persistence at end-of-life.

Binders used in the development of biodegradable radiative cooling paint require low solar spectrum absorptance and high sky window (8–13  $\mu\text{m}$ ) emissivity. The full spectrum refractive index and extinction coefficient of CA in UV, VIS, NIR, and MIR bands were not available in the literature; hence we characterize them in this work. To quantify this, an algorithm was developed to estimate the refractive index ( $n$ ) and extinction coefficient ( $\kappa$ ) of film samples, which serve as a thickness-independent indicator of optical properties. Starting with experimentally measured percent absorptance, reflectance, and transmittance, the corresponding coefficients are calculated via Look's equations.<sup>34</sup> With the absorption coefficient  $\alpha$ , the extinction coefficient  $\kappa$  can be found via

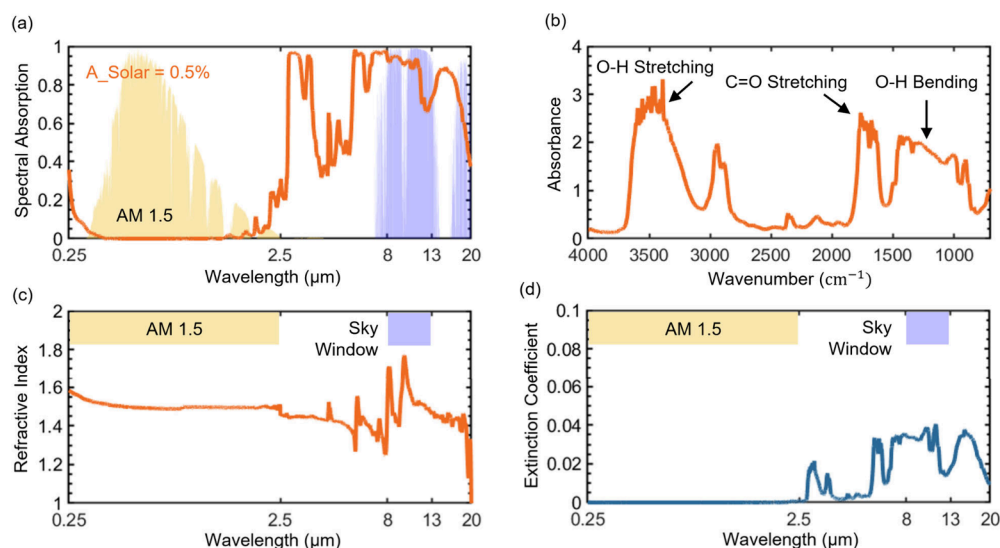
$$\kappa = \frac{\alpha\lambda}{4\pi}$$

where  $\lambda$  is the corresponding input wavelength in units of microns. Once  $\kappa$  is known, the refractive index can be found via

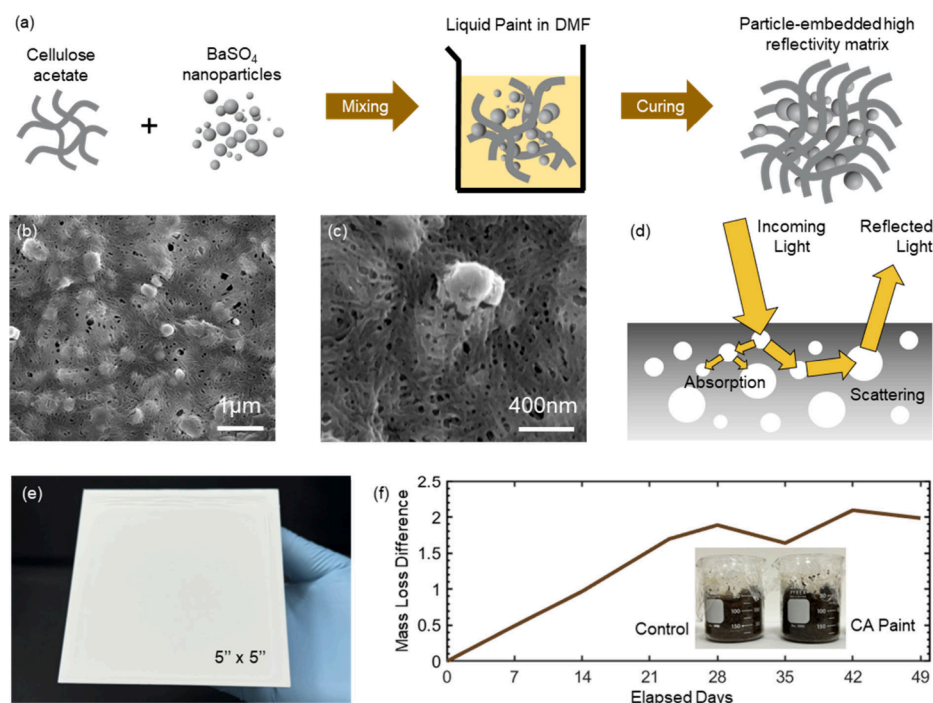
$$n = \frac{1 + R}{1 - R} + \left( \frac{4R}{(1 - R)^2} - \kappa^2 \right)^{0.5}$$

where  $R$  is the reflectance.

The design principle of radiative cooling paint seeks high refractive index, low extinction coefficient in the UV–visible–NIR region, and high extinction coefficient in the mid-IR region, especially around the 8–13  $\mu\text{m}$  “sky window”. Cellulose acetate (CA) exhibits low absorption in the visible spectrum (Figure 2a), as evidenced by its remarkably low



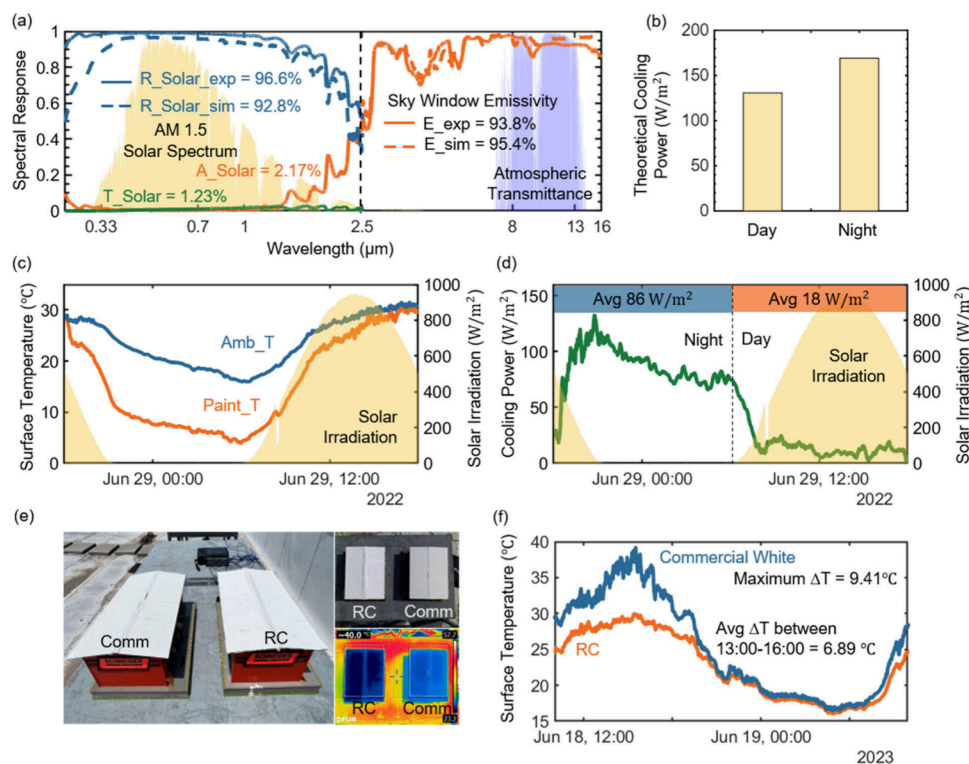
**Figure 2.** Binder selection. (A) Spectral absorptance and total solar absorption ( $A_{\text{solar}}$ ) of a pure cellulose acetate film as a function of wavelength in the AM1.5 solar region (reference air mass 1.5 spectra by NREL<sup>41</sup>) as well as the IR region, including the atmospheric transparency window (Atm.T). (B) IR region absorbance of a CA film as a function of wavenumber. (C, D) UV-vis-IR spectrum refractive index and extinction coefficient for a pure CA film.



**Figure 3.** Sample manufacturing and biodegradability testing. (A) Production process schematic of CA-based  $\text{BaSO}_4$  paint, including dissolution of binder, mixing of nanoparticles, and curing. (B, C) SEM images showcasing a web-like surface microstructure of a cured sample of  $\text{BaSO}_4$ -CA radiative cooling paint. (D) Schematic of the cross-sectional view of a cured sample featuring low absorption and high scattering effect, leading to high solar reflectance. (E) Cellulose acetate paint applied on an aluminum substrate. (F) Biodegradability testing of CA-based paint compared to its nonbiodegradable counterpart specifically an acrylic-based radiative cooling paint.

extinction coefficient and superior emissivity in the mid-infrared (MIR) region (Figure 2c,d) in comparison with the polyacrylic binder successfully used in past literature regarding radiative cooling paint (see Supplementary Note 1, a table of values for optical constants is also provided). Notably, Figure 2a also reveals that CA exhibits noticeable absorption in the ultraviolet (UV) region. This behavior is typical for organic polymers and represents a key limitation for relying solely on CA for solar reflectance. The presence of multiple absorption

peaks of CA is indicative of the various interatomic bonds, notably the O-H and C-O bonds, resonating at wavelengths within the sky window, contributing significantly to the high infrared (IR) emittance characteristic of CA (Figure 2b). Overall, CA exhibits a solar-spectrum optical response comparable to acrylic in the visible-NIR region while providing superior MIR emissivity, enabling effective rejection of heat through sunlight reflection and thermal emission to deep space, making it a suitable binder for radiative cooling. In



**Figure 4.** Optical characterization and outdoor testing. (A, B) Measured and simulated UV–vis–NIR spectral response of BaSO<sub>4</sub>–CA paint, along with corresponding theoretical cooling power. (C) Outdoor testing for subambient surface temperature and (D) night/daytime cooling power. (E) DSLR and thermal camera photographs of the testing apparatus (F) Temperature vs time between the CA–BaSO<sub>4</sub> radiative cooling paint and commercial white paint applied on the roofs of model buildings.

addition, CA is a biodegradable polymer known to be a nonirritant, heat resistant, nontoxic, and relatively less hygroscopic in nature. Hydroxypropyl cellulose acetate is an odorless, flavorless, transparent, stable, oil-resistant, nontoxic, and edible material with good film-forming properties. The biodegradability of CA has been extensively studied and widely acknowledged.<sup>35–39</sup> It is generally recognized that CA is readily biodegradable.<sup>40</sup>

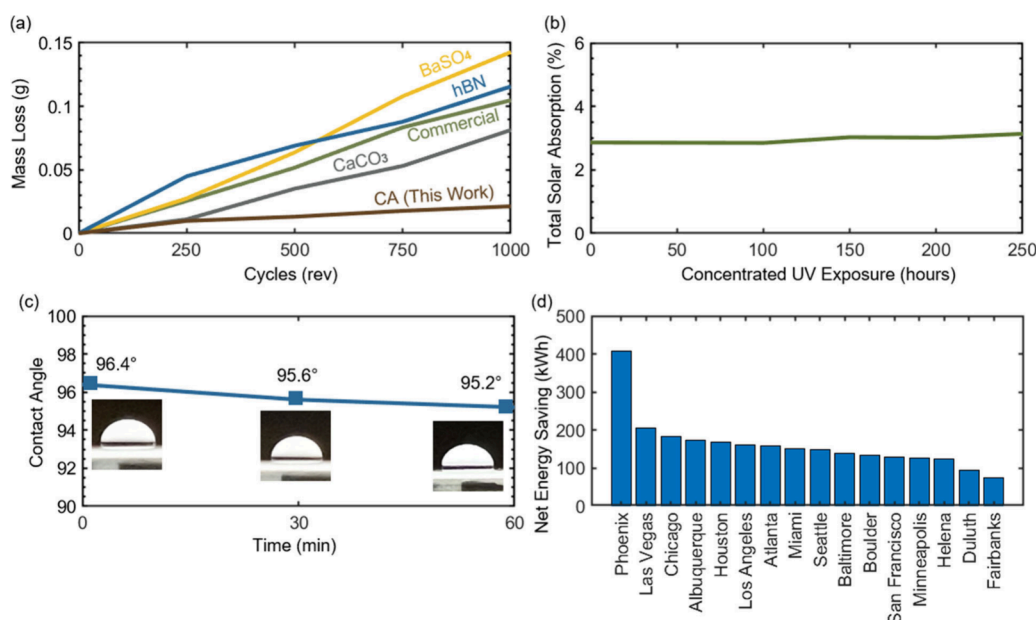
Previous studies have shown that CA alone functions as an effective radiative cooling material;<sup>42–44</sup> however, its intrinsic UV absorption limits its solar reflectance. Barium sulfate (BaSO<sub>4</sub>) nanoparticles, approximately 400 nm in diameter, exhibit high scattering properties in both the UV and NIR regions.<sup>10</sup> When included within the CA matrix, BaSO<sub>4</sub> effectively counteracts the inherent limitations of CA as a radiative cooling material.

To fabricate the CA-based radiative cooling paint, BaSO<sub>4</sub> nanoparticles and CA were mixed and sonicated in dimethylformamide. This sonication procedure ensures homogeneous distribution of particles within the solvent and prevents particle/cellulose fibers from clumping or sticking to the sidewalls. The resulting mixture had a viscous consistency and required at least 48 h to degas and fully cure (Figure 3a). The SEM image of a cured sample depicts an interesting web-like surface microarchitecture with BaSO<sub>4</sub> nanoparticles uniformly distributed and embedded in a matrix of CA binder (Figure 3b and 3c). A zoomed-in view of the paint surface shows an even distribution of nanopores between 20 and 100 nm in diameter (Figure 3d), along with BaSO<sub>4</sub> nanoparticles located near the surface. The paint effectively cures to a matrix-particle medium where the dominance of the scattering effect of nanoparticles in a porous matrix contributes

to high reflectance to incoming sunlight, while the CA binder, combined with the phonon resonance of BaSO<sub>4</sub> at 9 μm, ensures high emission within the sky window. Figure 3(e) shows a cured sample deposited on an aluminum substrate. The top surface is uniformly white, with a smooth and sturdy finish.

Figure 3(f) shows a comparison in biodegradability between a nondegradable sample of BaSO<sub>4</sub>–acrylic radiative cooling paint and a sample BaSO<sub>4</sub>–CA radiative cooling paint. With both samples fully submerged in compost, cellulose-based paint has shown complete degradation of the cellulose binder between 28 and 35 days of experiment, as the mass loss difference between the degradable (BaSO<sub>4</sub>–CA) and non-degrading (BaSO<sub>4</sub>–acrylic) sample has exceeded the initial mass of CA at the start of experiment. This observation is further reinforced as the steadily positive slope of the data is replaced by a plateauing, oscillating trend, suggesting the dominance of noise rather than data. It is important to note that BaSO<sub>4</sub> is a low-toxicity, biocompatible inorganic pigment and is not expected to biodegrade; therefore, this experiment does not demonstrate complete degradation of the entire paint composite. Rather, it demonstrates replacement of a persistent acrylic binder with a degradable cellulose acetate binder while retaining an inorganic optical pigment.

An opaque sample of CA–BaSO<sub>4</sub> radiative cooling paint achieves a high total solar reflectance of 96.6% and a high sky window emissivity of 0.938, which translates to a daytime theoretical cooling power of 130.7 W m<sup>−2</sup> and nighttime cooling power of 169.2 W m<sup>−2</sup> (Figure 4a,b). A Mie theory–Monte Carlo based algorithm<sup>45,46</sup> is used to simulate the spectral reflectance of BaSO<sub>4</sub>–CA paint, which yields a total solar reflectivity of 92.8%. The lower simulation results than



**Figure 5.** Practicality metrics. (a) Surface abrasion test conducted in accordance with ASTM D4060 for CA–BaSO<sub>4</sub> paint in comparison to several other paints. (b) UV degradation curve, characterized by change in solar absorption, of the CA–BaSO<sub>4</sub> sample under increasing exposure to concentrated UV light, up to 250 h. (c) Contact angle formed between a drop of DI water and the top surface of the paint, top coated with biocompatible PDMS polymer. (d) Aggregated annual energy savings (in kWh) as result of applying the paint to a ranch style home in cities across various US climates.

the experimental ones are due to the simulation's omission of scattering effects caused by micropores, as observed in the SEM images.

Radiative cooling capabilities were experimentally demonstrated through two apparatuses that measure the subambient capability and cooling power of paint samples, respectively (see [Methods](#)). [Figure 4\(c\)](#) shows clear 24-h subambient capability of CA paint with an average daytime (06:00–18:00) temperature difference of 5.1 °C below ambient, as well as an average nighttime (18:00–06:00) temperature difference of 10.9 °C below ambient. Among these, a peak temperature difference of 13.4 °C below ambient and cooling power of 132.4 W m<sup>−2</sup> were observed. This translates to an average cooling power of 17.8 W m<sup>−2</sup> during the day and 86.3 W m<sup>−2</sup> overnight ([Figure 4d](#)). While the subambient daytime cooling effect is primarily enabled by the paint's high solar reflectance, the paint's high nighttime cooling power stems from its high emissivity within the sky window, combined with clear, cloudless nights that provide unobstructed access to deep space.

To provide a comprehensive understanding of the advantages of BaSO<sub>4</sub>–CA radiative cooling paint over commercially available white paint, two identical single-story warehouse models were used as a testing apparatus. The roofs were painted with BaSO<sub>4</sub>–CA radiative cooling paint and commercial white paint, and the roofs' surface temperature were compared over a 24-h period in June 2023. This test differentiates itself from the previous one in that it does not attempt to eliminate the effect of ambient forced/free convection and conduction by suspending test samples in an isolated chamber. Instead, the paint samples were exposed to full ambient conditions under direct sunlight ([Figure 4e](#)), aiming to mimic the real-world conditions where buildings encounter various heat transfer mechanisms.

The model buildings are made mostly of cardboard and polymer, both with relatively low thermal conductivity. The

low conductivity of the model buildings not only mimics traditional insulation in walls and roofs but also helps minimize parasitic conduction from the metal table on which they are placed. The roof painted with biodegradable radiative cooling paint exhibited a significantly lower surface temperature during the day, reaching a maximum temperature difference of 9.4 °C and mean temperature difference of around 5 °C compared to the roof painted with commercial paint during peak daylight hours from 10:00 to 19:00 ([Figure 4f](#)). During the night, no significant temperature difference between the two groups is observed. This is expected, as the lack of solar irradiation deprives the highly reflective radiative cooling surface of any functionality and convection effects become dominant.

Cellulose acetate material is readily available and affordable at around \$1 per kilogram,<sup>47</sup> which is comparable to acrylic powders used in commercial products and other non-biodegradable radiative cooling paint products, with the added benefit of high strength and environmental friendliness. Combined with the cost of BaSO<sub>4</sub> nanoparticles at \$0.2 per kilogram,<sup>48</sup> it will cost about \$6 to cover 1 square meter of roof area. The mechanical durability of CA paint is tested with the sample subjected to abrasion profiles in accordance with the ASTM D4060<sup>49</sup> testing standard ([Figure 5a](#)). BaSO<sub>4</sub>–CA radiative cooling paint showed impressive resistance to surface abrasions, outperforming both acrylic-based radiative cooling paint and commercial exterior paint. This superior performance is attributed to the curing process of the BaSO<sub>4</sub>–CA paint, which results in a hard top shell with a high concentration of cellulose, creating a smooth and rigid surface capable of withstanding real-world environmental erosion. Note that the ASTM D4060 test is interpreted as a standardized comparative durability metric rather than a direct simulation of natural weathering. Because the tested sample was greater than 1 mm thick and abrasion was localized to a ring-shaped track, the measured material loss did not reduce the coating below the optically opaque thickness threshold.

Therefore, substantial changes in the overall solar reflectance are not expected from this level of localized abrasion.

The BaSO<sub>4</sub>–CA paint offers intrinsic advantages, such as low maintenance and a long service life. To detect its UV degradation capacity, BaSO<sub>4</sub>–CA paint was exposed to highly concentrated UV light in a weathering chamber for 250 consecutive hours. The chamber operates on ISO 11341:2004 standard with an accelerated aging ratio of around 6.4. Figure 5(b) shows the trend of total solar absorption vs weathering chamber exposure times as samples were taken out for measurements at specific intervals. Negligible UV degradation proved that the CA–BaSO<sub>4</sub> radiative cooling paint does not lose its effectiveness over long-term UV exposure.

The hydrophobicity of the BaSO<sub>4</sub>–CA radiative cooling paint can be achieved via a thin biocompatible PDMS topcoat (50–70 μm). The coated surface demonstrated sustained hydrophobicity, evidenced by a static contact angle of 96.4° with 0.01 cc of water, with a marginal decrease to 95.2° after 1 h under ambient indoor conditions (Figure 5c). This added hydrophobicity prevents premature biodegradation and fouling by giving the paint self-cleaning capabilities without significantly altering the radiative cooling capabilities. However, PDMS is a biocompatible, low-toxicity, and chemically inert plastic that is not considered biodegradable. While applying a layer of PDMS enhances its self-cleaning properties, it does come with the cost of increasing the nonbiodegradable content of this paint.

The radiative cooling capabilities of the BaSO<sub>4</sub>–CA radiative cooling paint are simulated in TRNSYS (Transient System Simulation Tool). We simulated the HVAC energy consumption from 12 AM, May 1, to 12 AM, October 1 (a typical “cooling season”), for a ranch style house<sup>50</sup> with its roof coated in BaSO<sub>4</sub>–CA radiative cooling paint, compared to the same house with conventional roof sidings. The HVAC system’s energy usage and efficiency (COP) is determined by the power consumption of TRNSYS’s built-in AC module (Type 921), which maintains an indoor set point temperature of 23 °C. Additional details on the TRNSYS model can be found in the Supporting Information. Net savings are calculated by subtracting the HVAC energy consumption of the painted house (experiment) from the consumption of the unpainted house (control). The results show that usage of CA–BaSO<sub>4</sub> radiative cooling paint yields better payback in hot and dry climates (Phoenix, AZ, 407.6 kWh) than in cooler, more humid climates (Los Angeles, CA, 167.4 kWh), consistent with previous literature on radiative cooling coatings.<sup>2</sup> Due to humid areas with increased rainfall and cloud cover, IR emission is blocked to deep space and therefore reduces the effect of radiative cooling. Despite this, simulation projects a cooling cost savings of \$47 throughout the cooling season, considering an average electricity cost of \$0.291/kWh in Los Angeles in March 2024, and a high cost saving of \$62.4 in hot, dry locations such as Phoenix, Arizona.<sup>51</sup> These findings confirmed that CA–BaSO<sub>4</sub> radiative cooling paint not only achieves substantial energy and cost savings in varying U.S. climates but holds promise for enhancing sustainability on a national scale.

This work contributes to the development of sustainable materials for thermal management by introducing a radiative cooling paint that is readily biodegradable, with 90% of its organic content decomposing within one year. The formulation incorporates a plant-based binder and a biocompatible filler, which together ensures that the product is nonirritating,

nontoxic, and odorless, and possesses self-cleaning capabilities. Our biodegradable radiative cooling paint features high solar reflectance (96.6%) and emissivity (0.938) within the atmospheric window, which, in practice, achieves peak cooling powers of 17.8 W/m<sup>2</sup> during the day and 86.3 W/m<sup>2</sup> at night. TRNSYS simulations highlighted its potential for significant energy savings, particularly in hot, dry climates such as Phoenix, AZ, where it can save over 400 kWh annually in a typical meteorological year. Furthermore, the paint’s mechanical durability, UV resistance, and cost-effectiveness validate its practical viability for widespread adoption.

## ■ ASSOCIATED CONTENT

### Data Availability Statement

Data needed to evaluate the conclusions in the paper are present in the manuscript or the Supporting Information. Additional background data are available from the corresponding authors upon request.

### Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.nanolett.6c01376>.

Additional experimental details and methods for paint fabrication, optical characterization, calculation of refractive index and extinction coefficient, compost biodegradation testing, outdoor cooling measurements, abrasion testing, and TRNSYS building-energy simulations; comparison of the spectral properties and optical constants of cellulose acetate and acrylic binders; evaluation of the effect of an optional PDMS topcoat on solar reflectance; and additional TRNSYS energy-consumption results (PDF)

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## Author Contributions

#Z.F. and Y.Z. contributed equally. X.R., T.L. Y.Z., and Z.F. conceived the study. Z.F. designed and performed the experiments, analyzed the results, and wrote the manuscript. Y.Z. performed the biodegradability study and some outdoor testing and captured SEM images. Z.G. performed the optical simulation. E.B. performed the cooling power simulation. W.-J.L. performed the UV resistance experiment. All authors contributed to discussions and revisions of the manuscript.

## Notes

The authors declare no competing financial interest.

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